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A MORE ACCURATE THERMAL NEUTRON COINCIDENCE COUNTING TECHNIQUE*

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INTRODUCTION

At the Los Alamos Scientific Laboratory, passive thermal-neutron coincidence counting is one of several techniques under investigation for the purpose of nondestructively assaying spontaneously fissioning material. Several thermalizing neutron detectors were constructed and their properties investigated. Such detectors have a cylindrical cavity, i.e., well, of about 6-in. diam x 20-in. long into which the sample is placed for counting. ^{3}He filled proportional counting tubes are placed at a fixed radius symmetrically about the well and are embedded in polyethylene (CH_2) moderating material. Designs studied include both one- and two-concentric rings of tubes. The well is generally lined with cadmium to 1) limit multiplication caused by thermalized neutrons being backscattered into the well, and 2) provide a system die-away time which is relatively independent of the sample. A typical two-ring detector is pictured in Fig. 1.

The amount of spontaneously fissioning material in the well is determined by comparing its coincidence count rate to that of a known mass, i.e., a standard. Since a sample's multiplication and moderation significantly affect the measured coincidence count rate per unit mass, an accurate comparison measurement demands that these effects be similar for the standard and unknown samples. This traditional approach presupposes a certain knowledge of the sample's matrix material, chemical composition, and geometry, which in fact are often unknown and furthermore necessitates the need for many standards. In such a context, this nondestructive assay (NDA) technique is somewhat expensive and cumbersome to use

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effectively. However, thermal-neutron coincidence counting becomes one of the more attractive NDA techniques from considerations of expense, accuracy, and versatility if a procedure can be found to account readily for multiplication and moderation effects within reasonable limits.

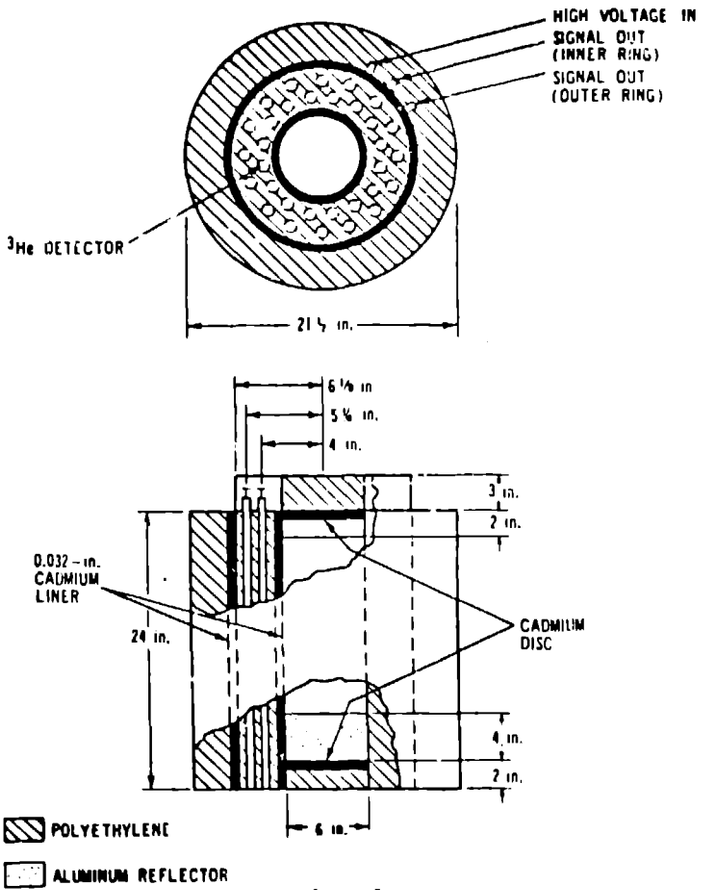

Fig. 1.
A two-ring thermal-neutron well-counter.

Very often NDA measurements must be made on samples of PuO_2 powder that are frequently contaminated with oxides or salts of light weight elements. Such contaminants have large (α,n) reaction cross sections, and the reaction product neutrons can cause significant multiplication. The object here is to investigate 1) the multiplication phenomena caused by both (α,n)- and spontaneous fission (SF)-neutrons associated with such powder samples, and 2) changes in neutron detection efficiency due to variations in neutron moderation among the samples. It is hoped that such work will lead to NDA neutron counting techniques that eliminate the measurement error introduced by failing to account properly for such phenomena.

This technique measures the amount of fertile material in the sample, which is assumed to consist of only isotopes of ^{239}Pu and ^{240}Pu. If there are other fertile isotopes present in addition to ^{240}Pu, then we measure the ^{240}Pu effective mass, M_{240}, which is defined to be the ^{240}Pu mass which will produce the same number of neutrons per second from spontaneous fissions as actually produced in all fertile isotopes in the mixed sample.

THEORY

General

We shall consider four sources of neutrons that contribute to the passive thermal-neutron singles count rate $\dot{T}$ defined as:

$$\dot{T} \equiv \dot{T}_{SF} + \dot{T}_{SM} + \dot{T}_{\alpha n} + \dot{T}_{IM} \quad . \tag{1}$$

The four contributors to $\dot{T}$ represent neutrons born by

a) spontaneous-fissions ($\dot{T}_{SF}$)
b) fissions induced by SF-neutrons and their progeny; these will be referred to as self-multiplication neutrons ($\dot{T}_{SM}$)
c) (α,n) reactions ($\dot{T}_{\alpha,n}$)
d) fissions induced by the (α,n)-neutrons and their progeny; these will be referred to as induced-multiplication neutrons ($\dot{T}_{IM}$)

The true coincidence count rate for neutrons created by fission process X must be related to the associated singles count rate. This can be expressed functionally as

$$\dot{n}_X = f_X(\dot{T}_X) \quad . \tag{2}$$

Since the measured true coincidence count rate $\dot{n}$ is the sum of the count rates of coincident pairs resulting from spontaneous-fissions ($\dot{n}_{SF}$), self-multiplications ($\dot{n}_{SM}$), and induced-multiplications ($\dot{n}_{IM}$), we can write

$$\dot{n} = \dot{n}_{SF} + \dot{n}_{SM} + \dot{n}_{IM} \tag{3}$$

However, the neutron detection efficiency is a function of its energy. Consequently, to make an accurate comparison measurement, it is imperative that our model modify $\dot{n}$ to

account for variations of neutron energy moderation among different samples. We note that neutrons contributing to true coincidence events are fission products and are therefore born with very nearly the same energy distribution (1). Therefore, we can reason that any variation among different samples of their energy distributions measured by the detector must be due to different moderating properties of the samples. We now define a modified coincidence count rate $\dot{N}$ whose magnitude is independent of the sample's moderation where

$$\dot{N} \equiv \frac{\dot{n}}{f(\text{sample's moderation})} \quad . \tag{4}$$

f(sample's moderation) is a function of the sample's moderation, i.e., an energy signature, which will be defined subsequently. Using Eq. 4, an expression for the modified total coincidence count rate analogous to Eq. 3 is defined as

$$\dot{N} \equiv \dot{N}_{SF} + \dot{N}_{SM} + \dot{N}_{IM} \quad . \tag{5}$$

The specific response rate SRR defined as

$$SRR \equiv \frac{\dot{N}_{SF}}{M_{240}} \tag{6}$$

is expected to be constant for a given detector. Using Eq. 5, the quantity $\dot{N}_{SF}$ that must be extracted from the coincidence count rate $\dot{N}$ in order to calculate SRR is given by

$$\dot{N}_{SF} = \dot{N} \cdot \left[\frac{\dot{N}_{SF}}{\dot{N}_{SF} + \dot{N}_{SM} + \dot{N}_{IM}} \right] \quad . \tag{7}$$

Defining the bracketed term in Eq. 7 to be the inverse of the correction ratio CR where

$$CR \equiv \frac{\dot{N}_{SF} + \dot{N}_{SM} + \dot{N}_{IM}}{\dot{N}_{SF}} \quad . \tag{8}$$

allows Eq. 7 to be rewritten as

$$\dot{N}_{SF} = \frac{\dot{N}}{CR} \quad . \tag{9}$$

Substituting Eq. 9 into Eq. 6 gives

$$SRR \equiv \frac{\dot{N}}{CR} \cdot \frac{1}{M_{240}} \quad . \tag{10}$$

Unless $\dot{N}$ and CR can be determined so that the coincidence count rate can be corrected for moderation and multiplication phenomena respectively, one cannot infer accurately the value of M_{240} in an unknown sample by comparing its response to that of a standard.

Multiplication Correction CR

Unfortunately a direct determination of CR is not possible since the quantities $\dot{N}_{SF}$, $\dot{N}_{SM}$, and $\dot{N}_{IM}$ are not measurable separately. As a result, there must be defined a function of some measurable quantity which is equivalent to CR in the sense that it will account adequately for modifications in the measured coincidence count rate due to multiplication phenomena. To do this it is necessary to identify the physical quantities which control these phenomena. This is done by writing more explicit definitions of the singles count rates summed in Eq. 1. The results indicate that these count rates and hence CR can differ among samples because of variations in 1) the probability P for generating random neutrons via the (α,n) reaction (where P is a function of the (α,n) reaction cross section $\sigma_{\alpha n}$ and the intimacy of the mixture μ of the matrix material and plutonium), 2) the energy E of the (α,n) reaction product neutrons which affects the multiplicities (2) associated with the induced multiplication and consequently the coincidence count rate $\dot{N}_{IM}$ (where E is controlled by μ, the (α,n) reaction Q value, and the neutron moderating properties of the sample), 3) the ^{240}Pu effective mass M_{240} which determines the number of SF-neutrons and the partial number of α-particles available to generate (α,n)- neutrons which can induce fissions, 4) the ^{239}Pu mass M_{239} which determines the partial number of α-particles available to generate (α,n)-neutrons which can induce fissions, 5) atom densities ρ_{239} and ρ_{240} of ^{239}Pu and ^{240}Pu respectively which

determine the probability that a neutron will induce a fission, 6) path length L of a neutron within a sample.

An attempt will be made to construct an empirical function of these variables that will describe relative values of CR by accounting for relative changes in $\dot{N}_{SM}/\dot{N}_{SF}$ and $\dot{N}_{IM}/\dot{N}_{SF}$ among different samples. To be useful, such a mathematical model of CR must result in a value of SIR that is constant over a reasonable range of values of these variables. Formalistically, CR can be written as a function of these variables where this function is defined as

$$CR \equiv f_{CR}(E, P, M_{239}, M_{240}, \rho_{239}, \rho_{240}, L) \qquad (11)$$

Experimental evidence that will be presented subsequently indicates that f_{CR} defined by Eq. 11 is separable, to a good degree of approximation, into the product of two independent functions, one of which depends only on the properties of the matrix material and the other on the properties of the plutonium. Therefore, since E and P are independent of M_{240}, M_{239}, ρ_{240}, ρ_{239}, and L, it is reasonable to assume that f_{CR} can be expressed as

$$CR \equiv f_{\alpha n}(E,P) \cdot f_{Pu}(M_{240}, M_{239}, \rho_{240}, \rho_{239}, L) \cdot \qquad (12)$$

Physically, $f_{\alpha n}(E,P)$ is that part of CR which accounts for variations in the induced fission rate due to different matrix materials. Its value is proportional to the probability that a random neutron of energy E will be generated by the (α,n) reaction on the matrix material. This quantity will be referred to as the matrix multiplication correction. We note that even among samples having the same neutron moderation, the matrix correction is not expected to be constant for a particular matrix material. This is due to the dependence of P on μ, the intimacy of the mixture of PuO_2 and the matrix material. The less intimate the mixture, the smaller will be the α-particle's energy prior to inducing the (α,n) reaction. This results in a smaller reaction product neutron energy E. Furthermore, in the presence of such a mixture the α-particle must travel farther to collide with a nucleus of the matrix material. This leads to an increased probability that the α-particle will be absorbed by a nucleus other than one of the matrix material thereby reducing the value of P. Thus we can expect perturbations about some average matrix correction $f_{\alpha n}$ due to variations in μ. The greater the intimacy of mixture, the larger will be P and E.

The factor f_{Pu} is proportional to the multiplication caused by SF-neutrons (which originate with ^{240}Pu) in the presence of plutonium densities ρ_{240} and ρ_{239}. In addition both M_{240} and M_{239} are sources of radioactive decay α-particles which are converted by the probability term $f_{\alpha n}$ to (α,n)-neutrons. Consequently f_{Pu} is also proportional to the multiplication caused by (α,n)-neutrons in the presence of ρ_{240} and ρ_{239}. The quantity f_{Pu} will be referred to as the plutonium multiplication correction.

Procedure to Calculate M_{240}

We will now sketch the manner by which the value of M_{240} can be determined for an unknown sample. Using Eq. 12 in Eq. 10 gives

$$SRR \equiv \frac{\dot{N}}{f_{\alpha n}(E,P) \cdot f_{Pu}(M_{240},M_{239},\rho_{240},\rho_{239},L)} \cdot \frac{1}{M_{240}} \quad . \tag{13}$$

where SRR is invariant and $f_{\alpha n}(E,P)$ is a function of the matrix material, its intimacy of mixture, and the sample's moderating property. Both SRR and $f_{\alpha n}$ are assumed to be measurable. Two simplifying assumptions are made which enable one to write $f_{Pu} \simeq f_{Pu}(M_{240})$. Defining the apparent specific response rate ASRR as

$$ASRR \equiv \frac{\dot{N}}{f_{Pu}(M_{240})} \cdot \frac{1}{M_{240}} \quad . \tag{14}$$

and using this definition in Eq. 13 allows us to write

$$ASRR = SRR \cdot f_{\alpha n}(E,P) \quad . \tag{15}$$

Once $f_{\alpha n}(E,P)$ and $f_{Pu}(M_{240})$ are defined and calculated using the results of measurements of standards, a calibration curve of ASRR vs $f_{\alpha n}(E,P)$ can be generated. The amount of ^{240}Pu in a sample for which $f_{\alpha n}(E,P)$ has been measured then can be determined using either the calibration curve or Eq. 15 to obtain ASRR and subsequently solving Eq. 14 for M_{240}.

Plutonium Correction f_{Pu} (M_{240}, M_{239}, ρ_{240}, ρ_{239}, L)

The function $f_{Pu}(M_{240}, M_{239}, \rho_{240}, \rho_{239}, L)$ cannot be modeled until we determine the sensitivity of the specific response rate to variations in these parameters. We now make the simplifying assumptions that 1) the PuO_2 powder samples are always in a constant geometry container and the fill heights are constant; therefore the density is proportional to the mass, and 2) all samples have the same isotopic abundances. Invoking these simplifying assumptions allows us to express f_{Pu} as a function only of M_{240} and L where the path length L is dependent on M_{240}. Furthermore, the empirical observation that the coincidence count rate varies non-linearly with the mass of plutonium suggests that we can write

$$f_{Pu}(M_{240}, M_{239}, \rho_{240}, \rho_{239}, L) = \frac{M_{240}^{X}}{M_{240}} \quad . \tag{16}$$

where the exponent X is an empirically determined constant. Substituting Eq. 16 into Eq. 14 gives

$$ASRR = \frac{\dot{N}}{M_{240}^{X}} \quad . \tag{17}$$

It is interesting to examine how restrictive are the simplifying assumptions of constant fill heights and isotopic abundances. Using the empirically determined value of X, one can estimate that variations in the fill height by ±20% affect the value of f_{Pu} by only ~±1%. A similar result is anticipated for variations in the isotopic abundance of ^{240}Pu.

Matrix Correction $f_{\alpha n}(E,P)$

The principal problem in this scheme to assay M_{240} is the identification and measurement of the function $f_{\alpha n}(E,P)$. This quantity must account correctly for changes in CR due to variations in E and P. We note that CR varies monotonically with E since the multiplicity and hence the coincidence count rate $\dot{N}_{IM}$ associated with the induced fission process varies monotonically with the incident neutron's energy (2). Furthermore, the value of CR also varies monotonically with P since the magnitude of $\dot{N}_{IM}$ varies monotonically with the number of random neutrons generated by the (α,n) reaction.

Fortunately a quantity that also varies monotonically with E and P can be defined and measured using a detector like that pictured in Fig. 1. A two-ring detector can provide information concerning the relative energy distribution of neutrons which enter the detector. This information is obtained by measuring the ratio of the neutron singles count rates in the outer and inner rings, $\dot{T}^O/\dot{T}^I$, where superscripts O and I refer to the outer and inner rings respectively. Using Eq. 1, this ratio can be written as

$$\frac{\dot{T}^O}{\dot{T}^I} = \frac{\dot{T}^O_{SF} + \dot{T}^O_{SM} + \dot{T}^O_{IM} + \dot{T}^O_{\alpha n}}{\dot{T}^I_{SF} + \dot{T}^I_{SM} + \dot{T}^I_{IM} + \dot{T}^I_{\alpha n}} \quad . \tag{18}$$

As previously mentioned, the SF-, SM-, and IM-neutrons are all fission products and to a good approximation their energy distributions at birth do not vary among different samples. However, the larger the energy E of the (α,n) neutron reaction product the harder will be the singles' spectrum and the larger will be the ratio $\dot{T}^O/\dot{T}^I$. Furthermore, inspection of Eq. 18 shows that if the average (α,n)-neutron energy $\bar{E}$ is greater than the average fission neutron energy $\bar{E}_f$,

$$\bar{E} > \bar{E}_f \quad , \tag{19}$$

then an increased value of P causes equal percentage increases in both $\dot{T}^O_{\alpha n}$ and $\dot{T}^I_{\alpha n}$ thereby increasing the ratio $\dot{T}^O/\dot{T}^I$. This effect is somewhat moderated by a concurrent increase in $\dot{T}^{O(I)}_{IM}$ where

$$\dot{T}^{O(I)}_{\alpha n} > \dot{T}^{O(I)}_{IM} \quad . \tag{20}$$

Therefore, both CR and $\dot{T}^O/\dot{T}^I$ have the same qualitative dependence on E and P.

Prompted by this qualitative similarity we assume that a function $f(\dot{T}^O/\dot{T}^I)$ can be defined so that

$$f^{O(I)}_{\alpha n}(E,P) \equiv f^{O(I)}(\dot{T}^O/\dot{T}^I) \tag{21}$$

for samples containing matrix materials that meet the criterion of Eq. 19 and whose (α,n) reaction Q values vary monotonically with expected values of P. We use the specific relation

$$f^{O(I)}(\dot{T}^O/\dot{T}^I) \equiv (\dot{T}^O/\dot{T}^I)^{\delta^{O(I)}} \tag{22}$$

to define $f_{\alpha n}^{O(I)}(E,P)$ in Eq. 15 where δ^O and δ^I are constants associated with the outer and inner rings respectively.

Detector Constants

Upon substituting Eq. 16 into Eq. 13 we can write the specific response rate as

$$SRR^{O(I)} = \frac{\dot{N}^{O(I)}}{f_{\alpha n}^{O(I)} \, M_{240}^X} \tag{23}$$

Using Eqs. 21 and 22, the above relation can be rewritten as

$$SRR^{O(I)} = \frac{\dot{N}^{O(I)}}{(\dot{T}^O/\dot{T}^I)^{\delta O(I)} \cdot M_{240}^X} \tag{24}$$

and equating the subsequent expressions of SRR for samples A and B yields

$$\left[\frac{(\dot{T}^O/\dot{T}^I)_A}{(\dot{T}^O/\dot{T}^I)_B}\right]^{\delta^{O(I)}} = \left[\frac{\dot{N}_A^{O(I)}}{\dot{N}_B^{O(I)}}\right] \cdot \left[\frac{(M_{240})_B}{(M_{240})_A}\right]^X \tag{25}$$

The exponents in Eq. 25 are obtained empirically in the following manner. A choice of δ^O determines values of X_i for several standard samples A_i comparisoned to standard B. A value of δ^O is found which minimizes the standard deviation calculated for the several values of X_i. Using this value

of δ^O and the counting data of several standards in Eq. 24 enable a least squares determination of SRR^O and X^O. This procedure is repeated using the counting data of the inner ring to determine δ^I, SRR^I, and X^I (which is very nearly equal to X^O). The final value of X is taken to be the average of X^O and X^I and the final values of SRR^O and SRR^I are determined using Eq. 24.

Using standards of PuO_2 powder mixed with different matrix materials, the singles and coincidence count rates were measured using a two-ring detector like that pictured in Fig. 1. The results of these measurements are listed in Table I. Since all of these samples were fired at the same temperature during their preparation, they are expected to have equivalent neutron moderation properties. In such a situation, there is no need to modify the measured coincidence count rate by a factor that accounts for varying moderation among these samples. We therefore define f(sample's moderation) $\equiv$ 1 in Eq. 4 and calculate the detector constants to be

$$\delta^I = 3.00 \tag{26-a}$$

$$\delta^O = 4.00 \tag{26-b}$$

$$X = 1.058 \tag{26-c}$$

$$(SRR^I)_{Av} = 24.94 \pm 2.24\% \tag{26-d}$$

$$(SRR^O)_{Av} = 19.62 \pm 1.07\% \tag{26-e}$$

Also listed in Table I are values of $ASRR^{O(I)}$ calculated using Eqs. 17 and 26-c. Pictured in Fig. 2 are plots of these values as a function of $f_{\alpha n}^{O(I)}(E,P)$ defined by Eqs. 21, 22, 26-a, and 26-b. The linear relation between $ASRR^{O(I)}$ and $(\dot{T}^O/\dot{T}^I)^{4(3)}$ indicates that the matrix correction defined by Eqs. 21 and 22 can be determined in the presence of an arbitrary mixture of these matrices.

Response of "Outer + Inner" Rings

We can improve the counting statistics and thereby reduce the measurement uncertainty by calculating a single response

TABLE I

COINCIDENCE COUNTING MEASUREMENT RESULTS[1]

SAMPLE ID	DOMINANT MATRIX MATERIAL	MASS OF Pu IN PuO_2 SAMPLE (grams)	EFFECTIVE MASS OF ^{240}Pu (grams)	TRUE COINCIDENCE COUNT RATES CORRECTED FOR BKGD & DEAD-TIME		"SINGLES" COUNT RATE RATIOS $\frac{i^0}{i^1}$	APPARENT SPECIFIC RESPONSE RATES		SPECIFIC RESPONSE RATES			SRR[B] CORRECTED FOR MODERATION[d]
				OUTER RING i^0	INNER RING i^1		OUTER[a] RING ASRR^0	INNER[a] RING ASSR^1	OUTER[b] RING SRR^0	INNER[b] RING SRR^1	BOTH[c] RINGS SRR^B	
STD 4	SiO_2	60.02	3.837	34.61	54.08	.8084	8.34	13.04	19.53	24.68	88.13	30.20
STD 5	MgO	60.03	3.838	39.03	60.83	.8407	9.61	14.66	19.27	24.67	87.55	31.88
STD 6	SiO_2	120.0	7.672	73.96	113.09	.8117	8.57	13.10	19.73	24.49	88.14	31.83
STD 7	MgO	120.0	7.672	85.25	132.19	.8424	9.87	15.31	19.61	25.61	90.03	31.62
STD 8	MgO	240.1	15.35	175.4	276.[illegible]7	.8367	9.76	15.38	19.90	26.25	91.88	30.82
STD 9	(none)	480.3	37.39	426.32	645.96	.8281	9.24	14.00	19.55	24.66	88.33	32.58
385	(none)	459.0	43.39	430.07	743.05	.8222	8.89	13.76	19.45	24.76	88.10	30.88
382	(none)	556.0	54.93	619.18	949.04	.8234	8.93	13.69	19.44	24.53	87.63	31.44
381	(none)	615.0	64.97	747.04	1141.92	.8229	9.03	15.80	19.69	24.77	88.63	32.01
447	(none)	779.0	81.36	981.29	1486.17	.8273	9.34	14.15	19.95	25.00	89.61	33.09

[1] No CH_2 placed about samples.

[a] Calculated using Eqs. 17, 26-c.

[b] Calculated using Eqs. 24, 26a-c.

[c] Calculated using Eq. 32.

[d] Calculated using Eqs. 32, 34, 35.

due to the counting data of both rings of a two-ring detector. To do this we define

$p^{O(I)}$ ≡ the average probability that one member of a pair of coincident neutrons will be detected by a counter in the outer (inner) ring.

If the average coincidence detection efficiency of SF-neutrons in the outer (inner) ring is defined as

$$\overline{\varepsilon^{O(I)}} \equiv \frac{SRR^{O(I)}}{a} \tag{27}$$

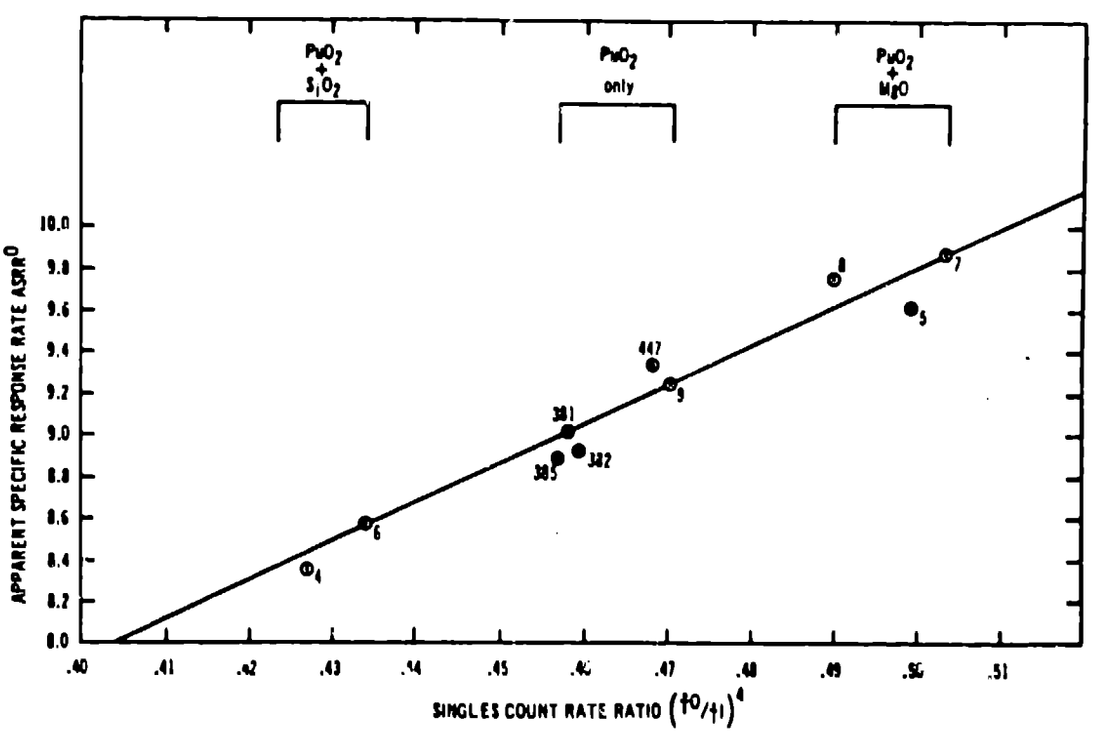

Fig. 2-a.
The apparent specific response rate of the outer ring versus the outer ring's matrix correction.

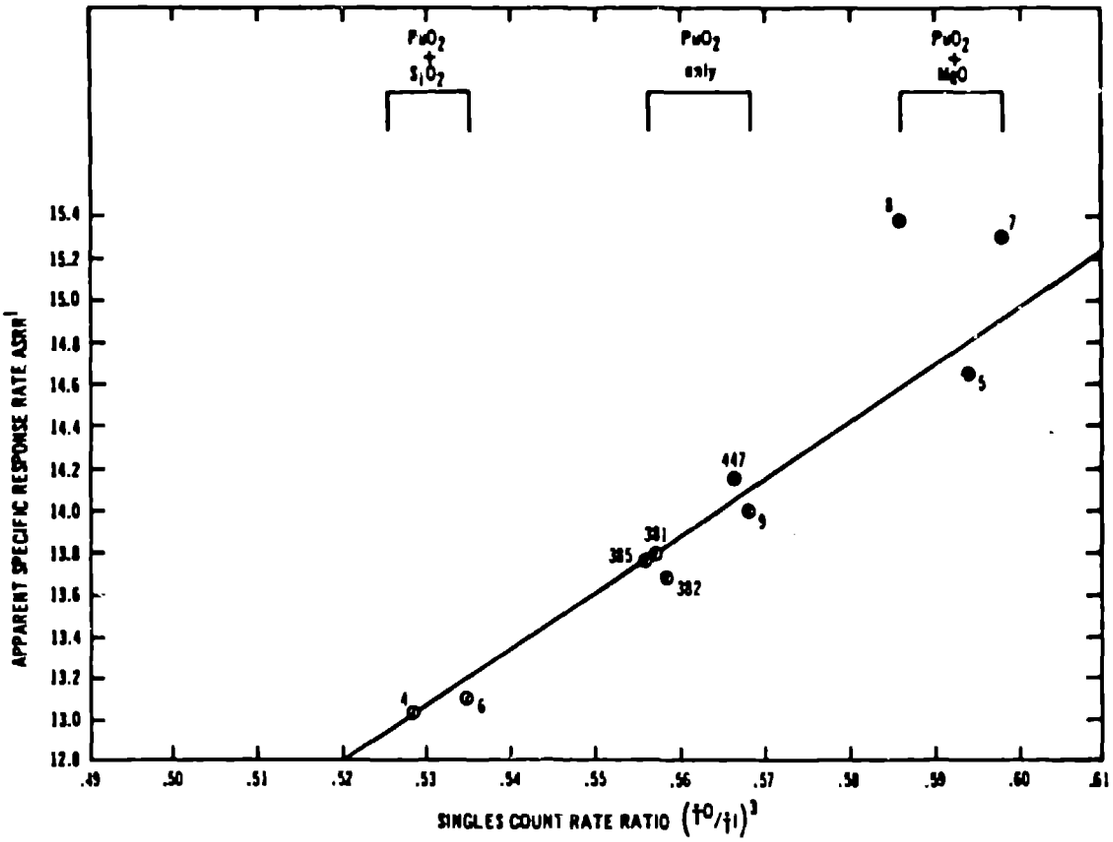

Fig. 2-b.
The apparent specific response rate of the inner ring versus the inner ring's matrix correction.

where a is the number of spontaneous fissions per s per gram of effective ^{240}Pu, then substituting Eqs. 24, 26a-c into Eq. 27 gives

$$\overline{\varepsilon^{O(I)}} = \frac{\dot{N}^{O(I)}}{a \cdot (\dot{T}^O/\dot{T}^I)^{4(3)} \cdot M_{240}^{1.058}} \tag{28}$$

Therefore, the probabilities $p^{O(I)}$ are

$$p^{O(I)} = \left[\frac{\dot{N}_{SF}^{O(I)}}{a \cdot (\dot{T}^O/\dot{T}^I)^{4(3)} \cdot M_{240}^{1.058}} \right]^{1/2} \tag{29}$$

The average efficiency of counting a pair of coincident neutrons by both rings of a two-ring detector can be written

$$\overline{\varepsilon^B} \cdot (p^I)^2 + (p^O)^2 + 2 \cdot p^I \cdot p^O \tag{30}$$

The specific response rate SRR^B which results from combining the responses of both rings is defined as

$$SRR^B \equiv \overline{\varepsilon^E} \cdot a \tag{31}$$

Substituting Eq. 29 into Eq. 30 and using the result in Eq. 31 allows us to write

$$SRR^B = \frac{\dfrac{\dot{N}^I}{(\dot{T}^O/\dot{T}^I)^3} + \dfrac{\dot{N}^O}{(\dot{T}^O/\dot{T}^I)^4} + 2\left[\dfrac{\dot{N}^I \cdot \dot{N}^O}{(\dot{T}^O/\dot{T}^I)^7}\right]^{1/2}}{M_{240}^{1.058}} \tag{32}$$

where the constant SRR^B is denoted as the calibration factor. Using the data of Table I, values of SRR^B are calculated and listed there.

Correction for Moderation

As was mentioned earlier in the discussion concerning Eq. 4, the neutrons contributing to true coincidence events are expected to be born with nearly the same energy distribution. Consequently, the ratio $\dot{n}^I/\dot{n}^O$ of the true coincidence count rates measured by the inner and outer rings of a two-ring detector should be a function of the moderating property of the sample's material. Referring to Eq. 4 we now define

$$f(\text{sample's moderation}) \equiv f_{MOD}(\dot{n}^I/\dot{n}^O) \tag{33}$$

Let us now examine the validity of this assumption.

In addition to the data already referred to in Table I, there is another data set listed in Table II for the same samples about which were placed the equivalent of 559 grams of CH_2. Values of SRR^B are calculated for this data using Eq. 24 and the detector constants in Eqs. 26a-c. The results are listed in Table II. As was done for the analogous calculation in Table I, we assume f(sample's moderation) $\equiv$ 1.

Pictured in Fig. 3 is a plot of these values of SRR^B vs $(\dot{n}^I/\dot{n}^O)^{2.4}$. We observe a grouping of the data. Thus it is reasonable to expect that a function of the ratio $\dot{n}^I/\dot{n}^O$ can be determined that will account for moderation effects in

TABLE II

COINCIDENCE COUNTING MEASUREMENT RESULTS[1]

SAMPLE ID	DOMINANT MATRIX MATERIAL	MASS OF Pu IN PuO_2 SAMPLE (grams)	EFFECTIVE MASS OF ^{240}Pu (grams)	TRUE COINCIDENCE COUNT RATES CORRECTED FOR BKGD & DEAD-TIME		"SINGLES" COUNT RATE RATIOS $\frac{\dot{T}^O}{\dot{T}^I}$	APPARENT SPECIFIC RESPONSE RATES		SPECIFIC RESPONSE RATES			SRR[B] CORRECTED FOR MODERATION[d] κ
				OUTER RING $\dot{N}^O$	INNER RING $\dot{N}^I$		OUTER[a] RING $ASRR^O$	INNER[a] RING $ASSR^I$	OUTER[b] RING SRR^O	INNER[b] RING SRR^I	BOTH[c] RINGS SRR^B	
STD 4	SiO_2	60.02	3.837	36.09	64.54	.7488	8.70	15.56	27.67	37.05	128.76	31.91
STD 5	MgO	60.03	3.838	42.53	76.23	.7646	10.25	18.37	27.05	38.04	129.25	31.85
STD 6	SiO_2	120.0	7.672	75.49	135.18	.7531	8.74	15.66	27.18	36.66	126.97	31.36
STD 7	MgO	120.0	7.672	89.12	159.19	.7824	10.32	18.44	27.55	38.50	131.19	32.60
STD 8	MgO	240.1	15.35	186.75	331.56	.7799	10.39	18.44	28.07	38.87	133.00	33.53
STD 9	(none)	480.3	37.39	440.72	781.08	.7686	9.55	16.93	27.38	37.30	128.59	32.56
385	(none)	459.0	43.39	497.22	894.15	.7666	9.21	16.56	26.66	36.76	126.03	30.82
382	(none)	556.0	54.93	651.46	1170.96	.7674	9.40	16.90	27.11	37.39	128.18	31.37
381	(none)	615.0	64.97	772.66	1380.41	.7690	9.34	16.68	26.69	36.67	125.93	31.28
447	(none)	779.0	81.36	1038.01	1870.26	.7731	9.88	17.43	27.67	37.72	130.00	33.32

[1] 559 grams CH_2 placed about samples

[a] Calculated using Eqs. 17, 26-c.

[b] Calculated using Eqs. 24, 26a-c.

[c] Calculated using Eq. 32.

[d] Calculated using Eqs. 32, 34, 35.

each sample. Consequently we define the modified coincidence count rates $\dot{N}^{O(I)}$ that appear in Eq. 5 to be

$$\dot{N}^{O(I)} \equiv \frac{\dot{n}^{O(I)}}{f_{MOD}(\dot{n}^{I}/\dot{n}^{O})} \tag{34}$$

where

$$f_{MOD}(\dot{n}^{I}/\dot{n}^{O}) \equiv (\dot{n}^{I}/\dot{n}^{O})^{2.4} \tag{35}$$

for the detector pictured in Fig. 1. Therefore, values of SRR^{B} calculated using Eqs. 32, 34, and 35, are independent of sample moderation. We denote these moderation-independent values of SRR^{B} as κ. Values of κ are calculated for the data presented in Tables I and II and also listed there.

The mean specific response rates with and without 559 g of CH_2 placed about the samples are calculated and listed in

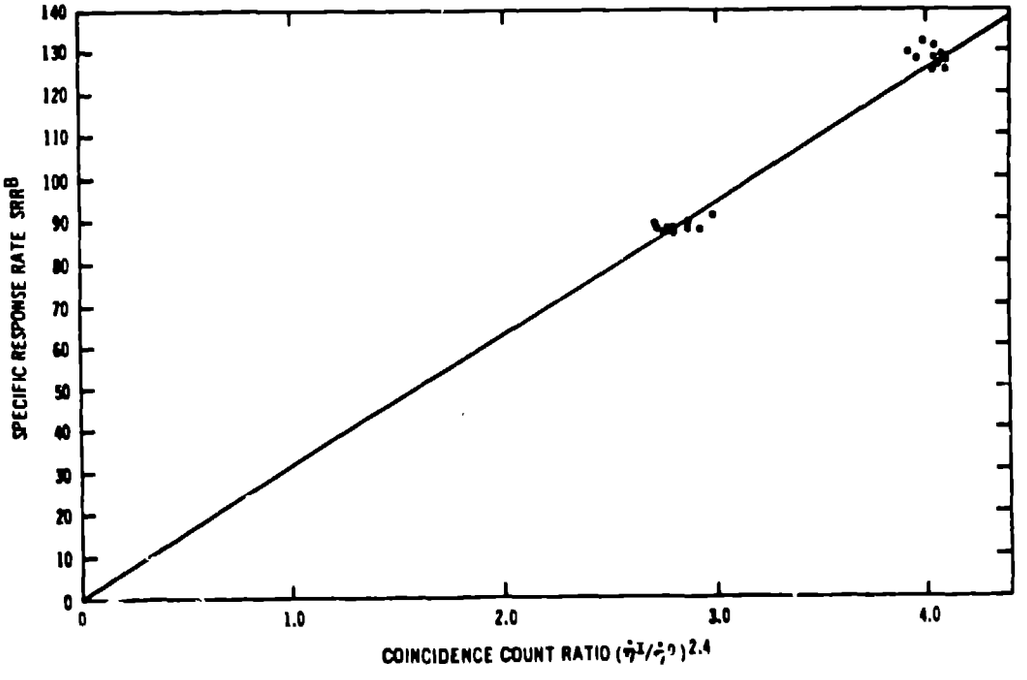

Fig. 3
The specific response rate SRR^{B} as a function of the coincidence count rate ring ratio.

Table III. The ratio of these mean specific response rates, for which no moderation correction has been made, is calculated as

$$\left[\frac{SRR^{B}_{Av}\ (559\ \text{grams of}\ CH_2)}{SRR^{B}_{Av}\ (0\ \text{grams of}\ CH_2)}\right] = 1.45 \pm 3.33\%$$

Similarly the ratio of the mean specific response rates corrected for moderation, which are also listed in Table III, is calculated as

$$\left[\frac{\kappa_{Av}\ (559\ \text{grams of}\ CH_2)}{\kappa_{Av}\ (0\ \text{grams of}\ CH_2)}\right] = 1.01 \pm 3.96\%$$

Thus we eliminate the 45% increase in specific response rates caused by the 559 grams of CH_2 moderating material. Furthermore, the mean value and associated standard deviation of the 20 specific response rates κ corrected for sample moderation, listed in the last columns of Tables I and II are calculated to be

$$\kappa_{Av} \pm \sigma_{\kappa_{Av}} = 31.84 \pm 2.73\%$$

This small standard deviation for samples having such grossly different moderating properties indicates the validity of correcting for moderation effects by using a function of the coincidence count rates $\dot{n}^I/\dot{n}^O$.

The Effective Mass of ^{240}Pu

The effective mass of ^{240}Pu in an unknown sample for which $\dot{n}^{O(I)}$ and $\dot{T}^{O(I)}$ have been measured can be calculated using Eqs. 32, 34, and 35. The final expression is

$$M_{240} = \exp\left\{\frac{\ell n\left(\dfrac{\dot{N}^I}{(\dot{T}^O/\dot{T}^I)^3} + \dfrac{\dot{N}^O}{(\dot{T}^O/\dot{T}^I)^4} + 2\cdot\left[\dfrac{\dot{N}^I\cdot\dot{N}^O}{(\dot{T}^O/\dot{T}^I)^7}\right]\right)^{1/2}}{SRR^{B}_{Av}}}{1.058}\right\} \quad (36)$$

TABLE III

MEAN VALUES OF SPECIFIC RESPONSE RATES

	NO POLYETHYLENE ABOUT SAMPLE		559 GRAMS POLYETHYLENE ABOUT SAMPLE	
	SRR^B (a)	κ (b)	SRR^B (a)	κ (b)
$\left(\begin{matrix}\text{SPECIFIC}\\ \text{RESPONSE}\\ \text{RATE}\end{matrix}\right)_{Av}$	88.8	31.6	128.8	32.1
$\sigma\left(\begin{matrix}\text{SPECIFIC}\\ \text{RESPONSE}\\ \text{RATE}\end{matrix}\right)_{Av}$	±1.5%	±2.7%	±1.7%	±2.8%

a) Calculated using Eq. 32.

b) Calculated using Eqs. 32, 34, 35.

where

$$\dot{N}^{O(I)} = \frac{\dot{n}^{O(I)}}{(\dot{n}^{I}/\dot{n}^{O})^{2.4}} \tag{37}$$

$$SRR^{B}_{Av} = \frac{\sum_{i=1}^{i_{max}} (SRR^{B})_{Std\ i}}{i_{max}} \tag{38}$$

and $(SRR^{B})_{Std\ i}$ is calculated using Eq. 32.

EXPERIMENTAL RESULTS

Using a two-ring well counter like that pictured in Fig. 1 and the theory described above, a standard deviation of less than 3% was obtained for the system's specific response rate. This figure reflects the expected accuracy for an assay of effective mass of ^{240}Pu. These results are based on the analyses of 20 samples of PuO_2 powders combined with different matrix materials among which 1) the probability or (α,n)-neutron induced fissions varied by 15%, 2) the multiplication varied by 28%, 3) a variable moderation effect changed the specific response rate by 45%, 4) plutonium densities varied by a factor of 6.5, and 5) masses of total plutonium varied from 60- to 779-grams.

CONCLUSIONS

Using passive thermal neutron coincidence counting techniques, the accuracy of nondestructive assays of fertile material can be improved significantly using a two-ring detector similar to that pictured in Fig. 1. It was shown how the use of a function of the coincidence count rate ring-ratio can provide a detector response rate that is independent of variations in neutron detection efficiency caused by varying sample moderation. Furthermore, the correction for multiplication caused by SF- and (α,n)-neutrons is shown to be separable into the product of a function of the effective mass of ^{240}Pu (i.e., plutonium correction) and a function of the (α,n) reaction probability (i.e., matrix correction). The

matrix correction is described by a function of the singles count rate ring-ratio. This correction factor is empirically observed to be identical for any combination of PuO_2 powder and matrix materials SiO_2 and MgO because of the similar relation of the (α,n)-Q value and (α,n)-reaction cross section among these matrix nuclei. However the matrix correction expression is expected to be different for matrix materials such as Na, Al, and/or Li. Nevertheless, it should be recognized that for comparison measurements among samples of similar matrix content, it is expected that some function of the singles count rate ring-ratio can be defined to account for variations in the matrix correction due to differences in the intimacy of mixture among the samples. Furthermore the magnitude of this singles count rate ring-ratio serves to identify the contaminant generating the (α,n)-neutrons. Such information is useful in process control.

REFERENCES

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